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13. ABSTRACT (Maximum 200 words)

This report results from a contract tasking Lebedev Physics Institute as follows: The contractor will continue to investigate and develop an experimental model of a portable optical frequency standard with reproducibility and accuracy of 10exp(-14) to 10exp (-15).

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TO EUROPEAN OFFICE OF AEROSPACE RESEARCH AND DEVELOPMENT

ATTENTION: Dr. Martin Stickley

Special Contract Program SPC-99-4023

Contract F61775-99-WE023

"Development of Diode-Pumped Color Center Laser with Methane Absorption Cell for Optical Frequency Standard (3.3 micron wavelength range)"

Final Report

<u>Part I</u>. Experiments with a CCL.

1. LD-based optical pumping system.

The main goal in this direction is as it was declared in the Project Statement to eliminate completely a Kr ion laser and substitute it by laser diodes (LD). An optical pumping system based on the powerful red (660 – 680 nm) LD for pumping a RbCl:Li (2.7 – 3.3 μm) color center laser (CCL) has been constructed. The pumping module is a rigid base where three LD and collimating optics are mounted. A number of optical schemes for convergence of laser beams from several LD were considered and tested. The scheme that we use now for pumping the CCL is shown in fig. 1. A microobjective is used for collimating the laser beam in the plane perpendicular to the junction (in diffraction limited direction). A cylindrical lens L_1 is used to obtain on the folding mirror M_1 of the CCL resonator (Fig. 2) a suitable spot size in non-diffraction limited direction (about 10 mm in our case). Cylindrical lenses L_2 and L_3 form a telescope (5:1) which reduces the beam diameter in an orthogonal direction thus permitting to combine in this direction beams from three lasers (Fig. 1b) on the folding mirror. The microobjective is a standard Newport F-L20B, AR-coated for λ=820 nm, that's why it's transmission at 670 nm (our pump wavelength) is only 82%. Proper AR coating will "save" at least 10% of pump power from each laser. All lenses (L_1, L_2, L_3) are AR-coated for red light. We use six LD which form two similar modules. The first consists of three broad band (100 μm) Coherent S-67-500C lasers with the following parameters:

P_1=370 mW, λ_1=671 nm,

P_2=290 mW, λ_2=667 nm,

P_3=415 mW, λ_3=674 nm

(output power has been measured after the microobjective). The total power of module-1 measured after the lens L_3 is about 1000 mW.

The other three LD form the second pumping module. These are SDL-7431 lasers (emitting area 1×250 μm):

P_4=400 mW, λ_4=677 nm,

P_5=425 mW, λ_5=674 nm,

P_6=200 mW, λ_6=675 nm.

The total power of module-2 is also about 1000 mW.

2. Operation of the CCL with LD pumping.

An optical scheme of the CCL cavity is presented in fig. 2. The pump radiation is coupled into the laser cavity via a dichroic mirror M_{IN}. This mirror must satisfy two requirements: high reflectivity at 3 μm and high transparency at pump wavelength (660-680 nm). We use mirrors made in Lebedev Institute optical workshop. Reflectivity at 3 μm depends on an incident angle and decreases from 99.5% to 96% when the angle changes in the range 0° - 35°. In the visible region the mirror works as a narrow-band filter - it has different transparency at different wavelengths. So in our case (for laser diodes operating at different wavelengths in the range 660-680 nm) we can achieve maximum transparency of the input mirror only for one LD. Taking this fact into account we can estimate total pump power reaching the crystal surface: it is about 800 mW from pumping module-1 and 600 mW from module-2, i.e. 1.4 W from all five LD. But it should be noted that this 1.4 W is not equal to 1.4 W of Kr^+ laser radiation because of lower efficiency of LD-pumping compared with Kr^+ laser pumping. This is a sequel of the specific spatial characteristics of the LD radiation. Powerful broad area LD generates a number of transverse modes and has non-diffraction divergence in the plane parallel to the junction. So it is difficult to achieve good fitting of the pump beam and the CCL resonator Gaussian TEM_{00} mode inside RbCl:Li crystal (the focused LD beam spot size inside the crystal is about 70×30 μm, CCL beam waist diameter ~70 μm). For the same reason efficiency of the 100 μm-band LD is higher than that of the 250 μm-band LD. Correspondingly we obtain three times lower pump power at threshold for pump module-1 LD in comparison with module-2.

Two schemes of the CCL pumping were used in the experiments. In the first one the radiation from two pump modules was coupled into the X-folded laser cavity from both ends via two dichroic mirrors (as shown in fig.2). In the second scheme the radiation from pump modules first was combined by means of a polarizing cubic beamsplitter. Thus only one input mirror was needed. Option depends on the quality of the input mirrors, optical density of the RbCl:Li crystal, the parameters of the pumping laser diodes. However in both cases all LD must be properly adjusted for obtaining CCL generation at the main spatial mode TEM_{00} (fig.3).

When pumped with maximum LD power the CCL in free running regime (without an intracavity etalon and absorption cell) was tuned over the range 2.75 – 3.31 μm with the aid of a diffraction grating (360 lines/mm, Al-coated) thus covering the R- and Q-branches of the methane ν_3-band.

3. Double mode CCL with an intracavity absorption cell.

The cavity configuration was chosen in such a way that mode competition was significantly weakened due to spatial hole burning of the population inversion in the active medium. An active crystal (RbCl:Li) is located in the center of the laser X-folded cavity (fig.2), so that $l = L/2$ (where L - the resonator length, l - the distance between the crystal and the nearest end mirror). Double mode operation with intermode spacing $\Delta\nu_{12}$ = 125 MHz (fig.4) was achieved with a help of a tunable intracavity etalon (ICE) with finesse ~2.7 and free spectral range ~7.5 GHz. The spectral width of the intermode beats was less than 2 kHz for an averaging time of 0.01 s, which was at the limit of the resolution of the spectral analyzer employed.

With the ICE the CCL can be tuned with the help of the diffraction grating from 2.8 to 3.28 μm. For each grating position by scanning only the ICE we can tune the CCL over the 7.5 GHz range with a resolution of 125 MHz corresponding to the CCL

intermode spacing. By means of such rough tuning and an external absorption cell which serves as a reference the CCL can be set at the chosen methane line. The low resolution Doppler and collisional broadened spectrum of the methane R(4) line is shown in fig. 5. Fine tuning without mode hopping over the 250 MHz range is achieved by synchronous scanning of the CCL and the ICE cavities.

The saturated dispersion resonances for the R(4)÷R(12) methane ν_3-band lines were observed in a scheme with an intracavity absorption cell. The cell length was 25 cm, the methane pressure was varied from 10 to 1 mTorr, the laser beam diameter inside the cell ~1.5 mm. The resonances were registered in the frequency of the intermode beat signal, which was detected by a fast (500 MHz) InAs photodetector. Fig.6 shows the saturated dispersion resonances for the R(9) line of the ν_3-band of methane ($\lambda = 3.21$ μm). The methane pressure in the cell was 7 mTorr. The F1(1) and F2(1) components of the R(9) line well resolved. The saturated dispersion resonance of the A1(1) component of the R(4) methane line ($\lambda = 3.26$ μm) obtained at the methane pressure of 1 mTorr is shown in fig.7. An estimate of the resonance width, deduced from the transmission peak of the marker cavity, yielded ~300 kHz. This value was determined primarily by the laser linewidth and by the transit-time broadening (120 kHz at T = 300 K and for laser beam diameter $2w = 1.5$ mm). Distortion of the dispersion resonance is associated with distortion of the signal in the narrowband registration system. More suitable broadband frequency converter is being designed now.

Applying of double-mode technique for detecting of the narrow resonances at the methane ν_3-band lines and using laser diodes for the CCL pumping allowed to increase by the order of magnitude the signal to noise ratio in comparison with a case of a singlemode CCL with a Kr^+ laser pumping.

Another important problem which is being solved is the problem of the intracavity losses. Now the LD-pumped CCL operates at $\lambda = 3.28$ μm just at threshold. (This wavelength corresponds to the R(2) methane line which we consider the most suitable for frequency standard.)Decreasing the total intracavity losses will facilitate stable operation at 3.28 μm. (From the other hand this way does not exclude the possibility of expanding the CCL tuning range by raising the pump power incident upon the crystal.) The problem of the intracavity losses is of great significance especially for a **transportable** optical frequency standard. Now an experimental set-up on the base of a He-Ne laser is made for measuring the losses of the optical elements in the 3 μm range. The results obtained at this installation are used by the specialists of an optical workshop in searching of the suitable low loss coatings for the mirrors and lenses used in the transportable He-Ne/CH4 optical frequency standard and in the CCL.

SubDoppler resonance of the A1(1) component of the methane R(6)-line recorded with the double mode CCL at methane pressure of 2 mTorr is shown in fig.8. Upper trace corresponds to the saturated dispersion resonance registrated in the intermode beat signal and lower trace corresponds to the saturated absorption resonance registrated in the intensity signal from both modes. The presence of the amplitude resonance in the net intensity of a double mode laser indicates that competition between the modes is significantly weakened. From the other hand the appearance of the detectable saturated absorption resonance points to the fact of total intracavity losses decreasing.

One of the main results of carried out experiments is solving of a problem of substitution of a Kr ion laser as a source of optical pumping. Now the CCL successfully operates under LD pumping practically over it's whole tuning range. And in all

experiments with the CCL on observation of the subDoppler resonances in methane (ν_3-band R-lines.) which were held last year laser diodes were used as a source of optical pumping.

Part II. Present performance of the transportable He-Ne/CH_4 optical frequency standards.

The second part of investigations was directed towards development and modification of a transportable optical frequency standard (TOFS) based on a He-Ne/CH_4 double-mode laser. These investigations on the one hand allow to realize a compact TOFS with stability and reproducibility on the level of 10^{-14}, and on the other hand they are very helpful for creating a new device based on a double-mode CCL/CH_4 laser.

One of the important parameters for practical use of the quantum frequency standards is a high level of a short and middle term frequency stability. The intense research is proceeding in this direction both for microwave and optical frequency standards. The physical principles of a double-mode methane based TOFS allow to reach high level of a short and middle term frequency stability in compact devices.

An example of the present level of frequency stability (sqrt of Allan variance), achieved at the Lebedev Institute with a new version of the double–mode He-Ne/CH_4 TOFS stabilized over resolved MHPS of $F_2^{(2)}$ component of the P(7) methane line (3.39 μm) with improved S/N ratio is demonstrated in fig.9 (curve2).

For comparison, the relative stability, determined for the earlier TOFS version during the absolute frequency measurements (with respect to the Cs/H-maser primary standard) at PTB (Braunschweig, Germany) in 1997 is shown also - a curve 1. The curve 1 indicates that for sample times $\tau > 10^3$ s the relative stability exceeds $2*10^{-14}$ but for $\tau < 10$ s the real performance is not clear.

Present measurements of the short and middle term frequency stability were carried out by comparison of two independent TOFS devices. The curve 2 clearly shows that the TOFS stability exceeds the H-maser performance more than one order for sample times $\tau < 1$ s. For longer sample times ($\tau = 100 - 1000$ s) the stability practically coincides with H-maser stability.

The development of the double-mode methane based CCL should allow of 10-50 times increase of the relative stability (up to $10^{-15} / \sqrt{\tau}$ for $\tau = 1$- 10^3 s) thus reaching the performance of the best stationary experimental setups.

The results of the experiments were reported at the 8th International Laser Physics Workshop Lphys'99 (Budapest, July 2-6, 1999) and at the Joint Meeting EFTF-IEEE (Besanson, France, 13-16 April, 1999), and were published in:

1. O.Acef, A. Clairon, G.D.Rovera, L.Hilico, G.Kramer, B.Liipphardt, A.Shelkovnikov,E.Kovalchuk, E.Petrukhbin, D.Tyurikov, M.Petrovskiy, M.Gubin, R.Felder, P.Gill, S Lea, "Absolute frequency measurements with a set of transportable methane optical frequency standards" , in Proceedings of 1999 Joint Meeting EFTF-IEEE IFCS, 13-16 April 1999, Besanson, France. IEEE Catalog #99CH36313, Library of Congres #87-654207, pp.742-745.

2. M.Gubin, A. Shelkovnikov, E.Kovalchuk, D.Krylova, E.Petrukhin, D.Tyurikov, "Present performance of the transportable He-Ne/CH4 frequency standards", in Proceedings of 1999 Joint Meeting EFTF-IEEE IFCS, 13-16 April 1999, Besanson, France. IEEE Catalog #99CH36313, Library of Congres #87-654207, pp.710-713.
3. M.Gubin, A.Kireev, E.Koval'chuk, D.Tyurikov, "Two-mode diode-pumped color center laser with an intracavity absorbing cell", Quantum Electron. **29**, p.1027 (1999)

In Conclusion. New horizons for a compact midinfrared - to- microwave frequency bridge.

The absence of convenient instruments for laser frequency measurement, differ from conventional optical synthesis chains [1] in terms of reliability, simplicity of operation and compactness, have restrained for a long time wide application of high accurate OFS, (especially at visible – UV range of spectra). One of the most inconveniences of conventional optical frequency synthesis chain is connected with necessity of usage a variety of different type of oscillators spreaded at the range 10^{10}-10^{14} Hz.

The new effective approach, introducing the measuring only frequency differences between the lasers instead of measuring the optical frequency starting from microwave range (like in conventional optical synthesis chains), was proposed at the beginning of 90-th [2]. The new scheme for measure optical frequencies uses only optical means: lasers and nonlinear crystals. As in case of conventional chains the new scheme also consists of several intermediate stages before reaching the microwave range.

In order to reduce the number of the intermediate stages and make the scheme really practical, the devices called as "optical frequency comb generators" were proposed [3]. The optical frequency comb generator is based on F-P resonator combined with electrooptic modulator, driven by microwave standard. They can generate a broad band of equally spaced optical frequency components (frequency comb) separated by the microwave modulation frequency. Optical coatings and electrooptic modulator parameters limit the optical frequency range covered by such device typically to a level of several THz, i.e. (5-10)% of the optical range frequencies.

Really revolutionary step for *dramatic widening* the optical frequency comb range was made in last two years by realizing a Ti : Sapphire laser ($\lambda = 0{,}85$ μm) continuously emitting coherent sequence of the phasematched ultra-short (10 – 5 fs) pulses [4].

Repetition rate of the fs-pulses (~10^9 MHz) is driven by microwave source and Fourier spectrum of these phasematched fs-pulses represents an optical frequency comb of equally spaced (with the same microwave frequency interval) components. In such manner the microwave source determines the accuracy of the comb spacing as well as the interval between extreme "red" and "blue" components of the fs -comb. The optical wavelength range covered by this type of fs-comb recently reached 532nm – 1,06 μm interval (!) [5]. It means that the reliable, compact, "one-step" optical-microwave frequency bridge is practically realized, thus opening the way to highly precise OFS.

The frequency of a methane stabilized OFS can be measured immediately as the 88 THz interval corresponding to the CH_4 transition is with an excess covered by the 300 THz comb span obtained in [6]. A potential compactness of the "88THz to 1 GHz fs – bridge" could be considered as very high, keeping in mind the fiber optics based fs-

lasers created in GB by Dr. Sillberg group. The power consumption for 18 hours of operation for this fs laser system is maintained by three AA size batteries only.

References:

1. G.Kramer, B.Lipphardt, U.Huebner, "Phase coherence in optical frequency measuerment and synthesis" in Proceedings of the 1999 Joint Meeting EFTF-IEEE IFCS, 13-16 April 1999, Besanson, France, IEEE Catalog #99CH36313, Library of Congres #87-654207, pp.651-654.
2. H.R.Telle, D.Meschede, and T.W.Hansch, "Realization of a new concept for visible frequency division: Phase locking of harmonic and sum frequencies, "Opt. Lett., **15**, pp.532-534, (1990).
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4. Th.Udem, J.Reichert, R.Holzwarth, and T.W.Hansch , Optics Lett., **24**, pp.881- 883 (1999)
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6. S.A.Diddams, D.J.Jones, Jun Ye et al, "Direct link between microwave and optical frequencies with a 300THz femtosecond laser comb", Phys. Rev. Lett., **84**, pp.5102-5105 (2000).

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16 June 2000

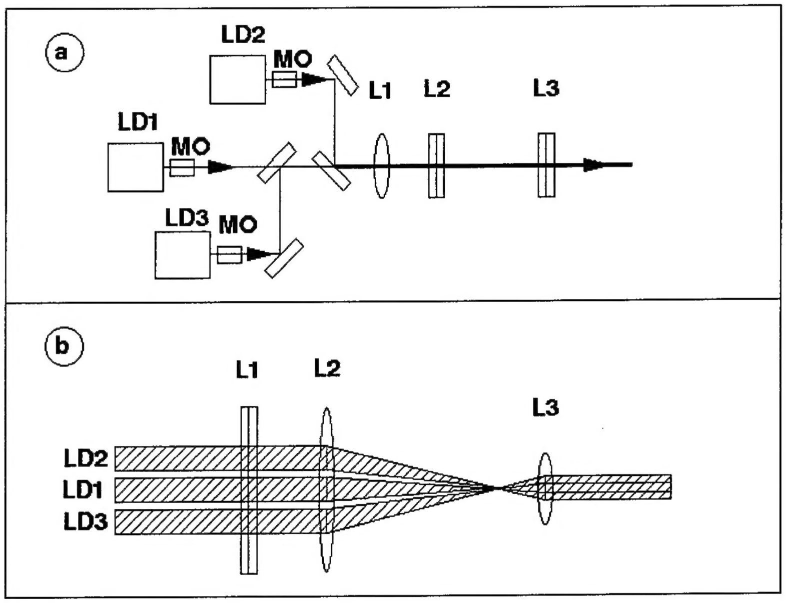

Fig.1. Optical scheme of the LD-based pumping system.
a) P - plane, b) S - plane.
LD_1, LD_2, LD_3 - laser diodes; MO - microobjective;
L_1, L_2, L_3 - cylindrical len9ses.

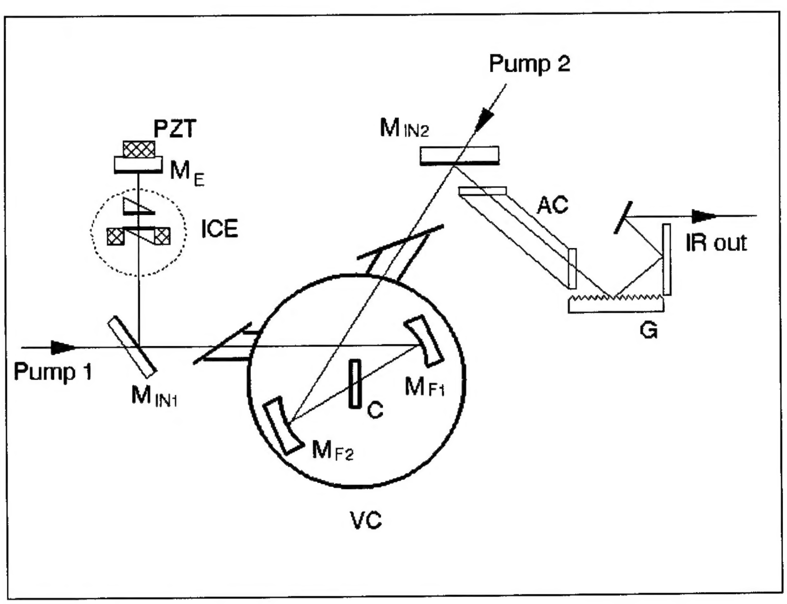

Fig.2. Optical scheme of the LD-pumped CCL.
C - RbCl:Li crystal; M_{F1}, M_{F2} - folding mirrors; M_{IN1}, M_{IN2} - input mirrors; ICE - tunable intracavity etalon; AC - internal absorption cell; G - diffraction grating; M_E - end mirror on PZT; VC - vacuum cryostat.

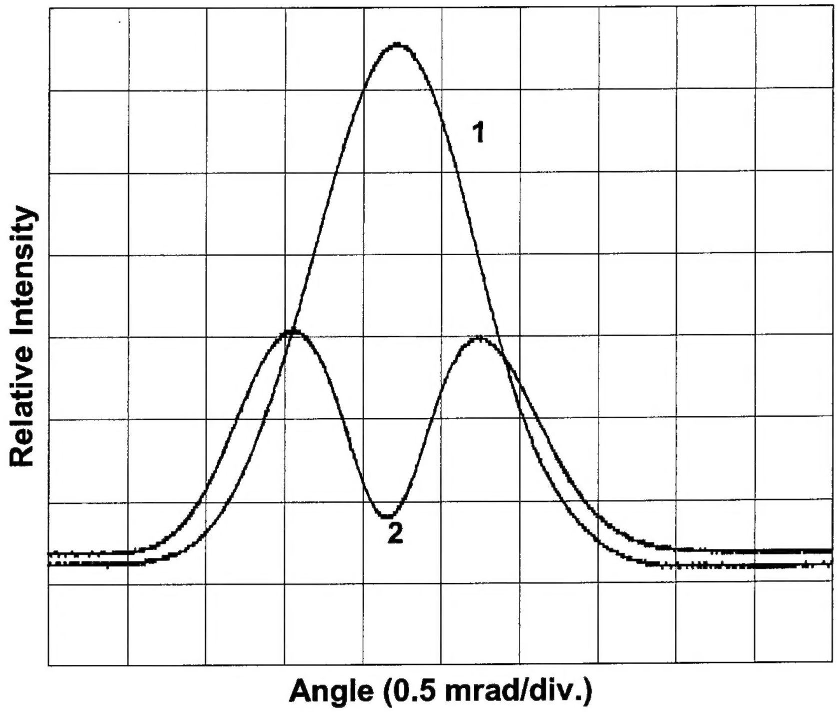

Fig.3. Far-field spatial distribution of the LD-pumped CCL. 1 - the main Gaussian mode, 2 - transverse mode.

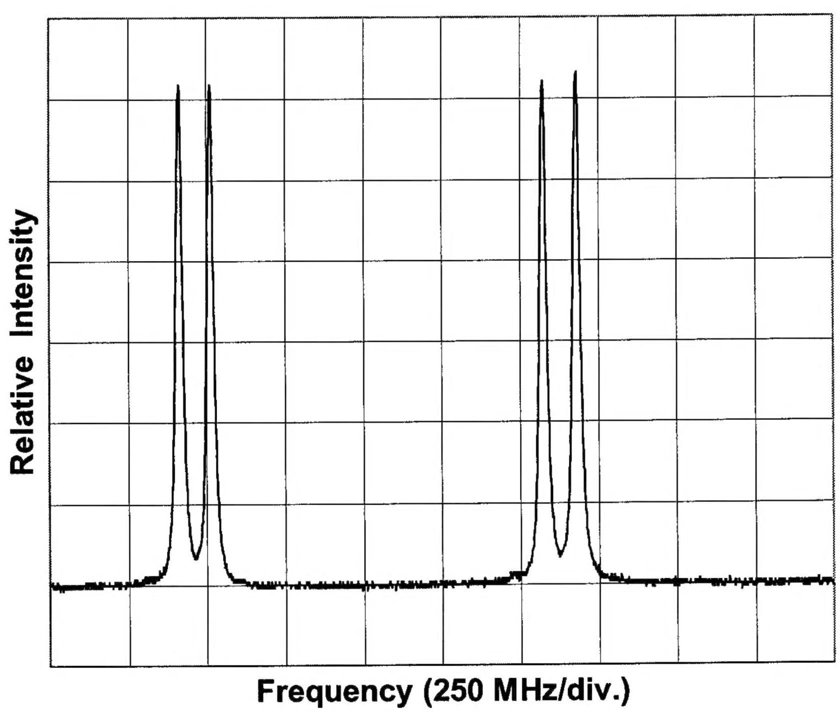

Fig.4. Transmission resonances of a confocal scanning interferometer.

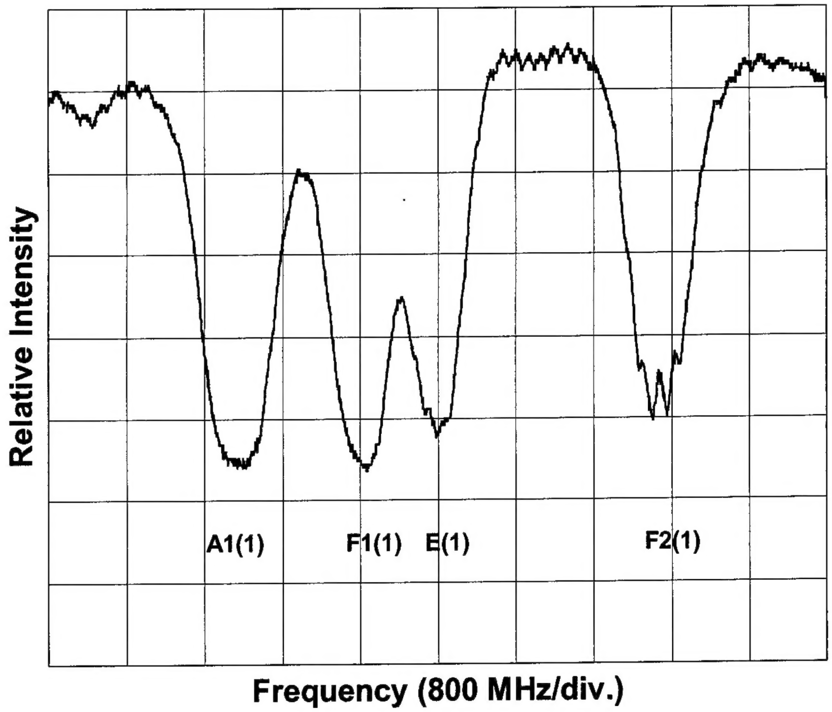

Fig.5. Low resolution spectrum of the methane R(4) line, external absorption cell (cell length 7 cm, pressure ~ 5 Torr CH_4 + 30 Torr air).

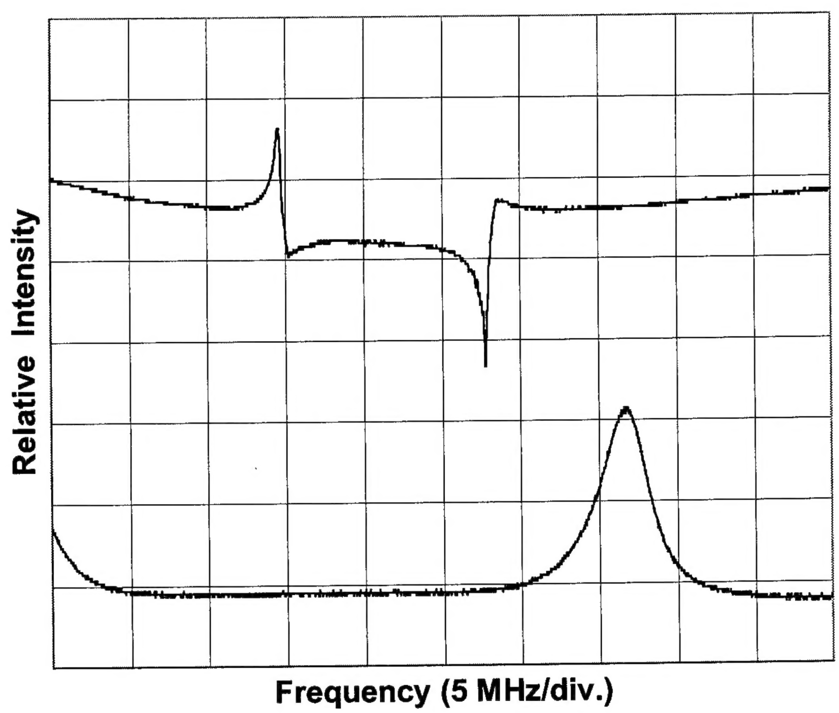

Fig.6. Saturated dispersion resonances of the F1(1) and F2(1) components of the methane ν_3-band R(9)-line (λ=3.21 μm).
Lower trace: marker cavity signal.

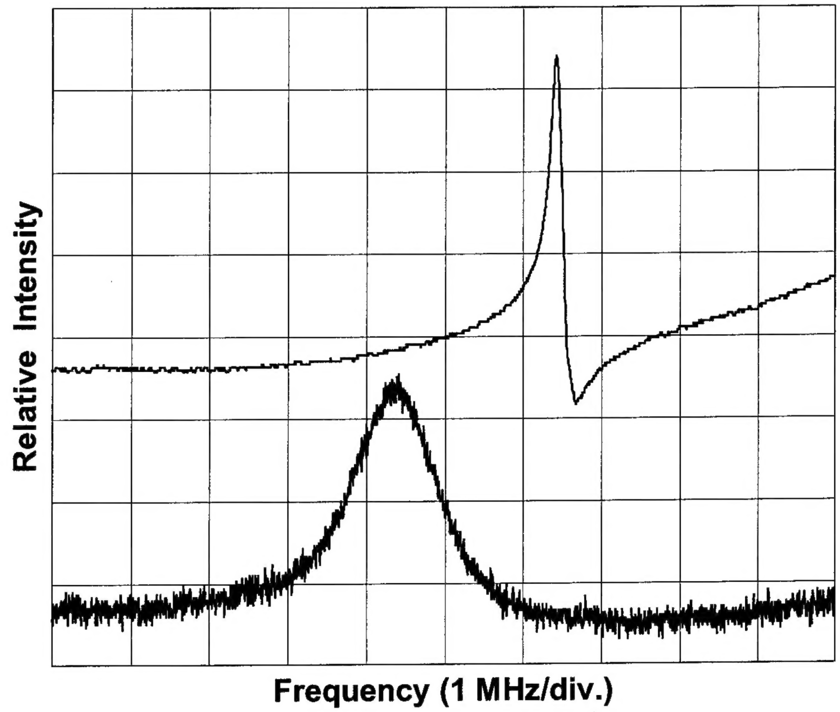

Fig.7. Saturated dispersion resonance of the A1(1) component of the methane ν_3-band R(4)-line (λ=3.26 μm).
Lower trace: marker cavity signal.

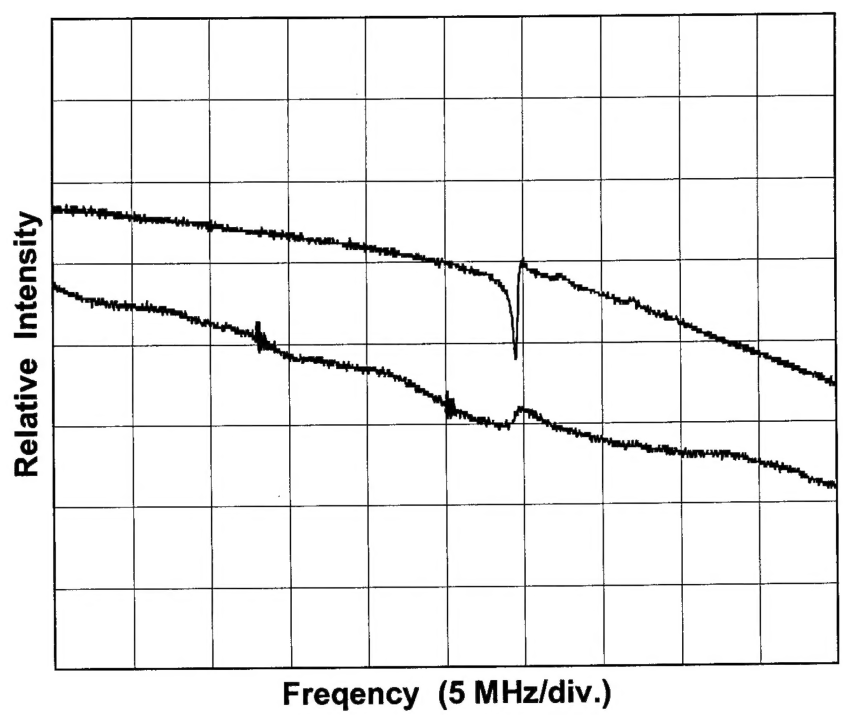

Fig.8. SubDoppler resonance of the A1(1) component of the methane ν_3-band R(6)-line (λ=3.24 μm).
Upper trace: saturated dispersion resonance,
lower trace: saturated absorption resonance.

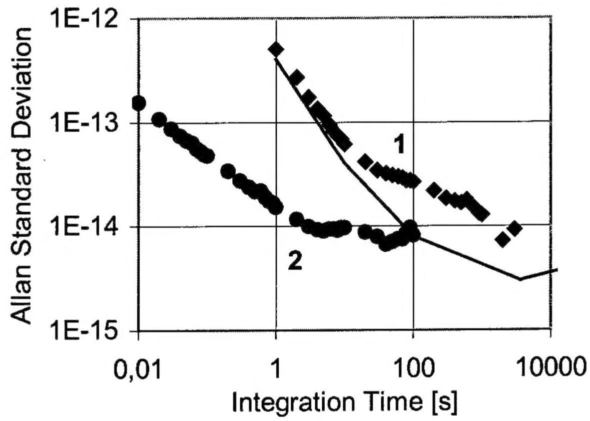

Fig.9. Allan standard deviation of the double-mode He-Ne/CH_4 TOFS measured at the PTB radio-optical frequency chain.
1 – earlier TOFS version (Nov.'97),
2 – new TOFS version (99),
Solid line - the PTB chain/H-maser accuracy limit.